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(19) (CA) APPLICATION FOR CANADIAN PATENT (12)

(54) N-Hydroxy-N-Phenylcarboxamides, Their Preparation and Compositions Containing Them for Controlling Harmful Fungi

5,075,8/4]

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(30) (DE) P 42 31 518.2 1992/09/21

(57) 7 Claims

Notice: This application is as filed and may therefore contain an incomplete specification.

Industrie Canada Industry Canada

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N-Hydroxy-N-phenylcarboxamides, their manufacture, and agents containing them for for combatting injurious fungi

#### 5 ABSTRACT OF THE DISCLOSURE:

1. N-Hydroxy-N-phenylcarboxamides of the formula I

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15 where:

> is substituted or unsubstituted alkyl, alkoxy, alkenyl, alkenyloxy, alkynyl, alkynyloxy, cycloalkyl, cycloalkenyl, cycloalkyloxy, cycloalkenyloxy or phenyl;

20 is one of the radicals A1 to A7

25 **A3** A4 A2 A1 30 Аб **A7** 

where

is -CH<sub>2</sub>-, -S-, -SO- or -SO<sub>2</sub>-; Х is -0- or -S-; 40  $R^2$ ,  $R^4$ ,  $R^5$  and  $R^7$  are halogen, alkyl or haloalkyl; R3 and R6 are hydrogen, halogen or alkyl; is 1 or 2;

45 methods of manufacturing them, agents containing them, and their use for combatting injurious fungi.

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#### N-HYDROXY-N-PHENYLCARBOXAMIDES, THEIR PREPARATION AND COMPOSITIONS CONTAINING THEM FOR CONTROLLING HARMFUL FUNGI

The present invention relates to N-hydroxy-5 N-phenylcarboxamides of the formula I

where the substituents have the following meanings:

 $C_2-C_{12}$ -alkoxy, C<sub>1</sub>-C<sub>12</sub>-alkenyl, C,-C12-alkyl, R  $C_3-C_{12}$ -alkenyloxy,  $C_3-C_6$ -alkynyl,  $C_3-C_6$ -alkynyloxy, where these groups can be partially or completely halogenated;  $C_3-C_7$ -cycloalkyl,  $C_4-C_7$ -cycloalkenyl,  $C_3-C_7$ -cycloalkoxy or  $C_4-C_7$ -cycloalkenyloxy, where these rings can carry one to 3 C1-C4-alkyls; phenyl which can carry one to five halogen atoms and/or one to three of the following radicals:  $C_1-C_4$ -alkyl, C<sub>1</sub>-C<sub>4</sub>-haloalkoxy,  $C_1-C_4$ -alkoxy, C,-C,-haloalkyl, C1-C4-alkylthio or C1-C4-haloalkylthio; is a cyclic radical from the group consisting of the

where the substituents have the following meanings:

20 X is -CH<sub>2</sub>-, -S-, -SO- or -SO<sub>2</sub>-;

formulae Al to A7:

y is -0- or -S-'

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 $R^1$ ,  $R^2$ ,  $R^4$ ,  $R^5$  and  $R^7$  are halogen,  $C_1-C_4$ -alkyl or

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C<sub>1</sub>-C<sub>4</sub>-haloalkyl;

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R<sup>3</sup> and R<sup>6</sup> are hydrogen, halogen or C<sub>1</sub>-C<sub>4</sub>-alkyl;

is 1 or 2, where the radicals  $R^3$  can be different if the value of n is 2.

The invention additionally relates to the preparation of these compounds, compositions containing them and their use for controlling harmful fungi, in particular Botrytis.

N-(2-Chlorophenyl)-2-chloronicotinamide is known 10 from the literature as a fungicidal active compound (DE-A 2 417 216).

It is an object of the present invention to provide novel fungicidally active compounds having an improved spectrum of action.

We have found that this object is achieved by the compounds I defined at the beginning.

We have additionally found processes for preparing these compounds, compositions containing them and their use for controlling harmful fungi.

The compounds I are in general obtained by reacting a carboxylic acid halide of the formula II in a manner known per se (eg. J. March, Advanced Organic Chemistry, 2nd Ed., 1977, 382 ff., McGraw-Hill) with an N-hydroxyaniline of the formula III in the presence of a base.

The radical Hal in the formula II is a halogen such as chlorine, bromine or iodine, in particular chlorine or bromine.

This reaction is customarily carried out at 30 temperatures from  $-20\,^{\circ}\text{C}$  to  $100\,^{\circ}\text{C}$ , preferably  $-10\,^{\circ}\text{C}$  to

50°C.

Suitable solvents are:

aliphatic hydrocarbons such as pentane, hexane, cyclohexane and petroleum ether, aromatic hydrocarbons such as
toluene, o-, m- and p-xylene, halogenated hydrocarbons
such as dichloromethane, chloroform and chlorobenzene,
ethers such as diethyl ether, diisopropyl ether, tertbutyl methyl ether, dioxane, anisole and tetrahydrofuran,
nitriles such as acetonitrile and propionitrile, ketones
such as acetone, methyl ethyl ketone, diethyl ketone and
tert-butyl methyl ketone, alcohols such as methanol,
ethanol, n-propanol, isopropanol, n-butanol and tertbutanol, and also dimethyl sulfoxide and dimethylformamide, particularly preferably toluene, xylene and
tetrahydrofuran.

Mixtures of the solvents mentioned can also be used.

Suitable bases are generally inorganic compounds such as alkali metal and alkaline earth metal hydroxides 20 such as lithium hydroxide, sodium hydroxide, potassium hydroxide and calcium hydroxide, alkali metal and alkaline earth metal oxides such as lithium oxide, sodium oxide, calcium oxide and magnesium oxide, alkali metal and alkaline earth metal hydrides such as lithium 25 hydride, sodium hydride, potassium hydride and calcium hydride, alkali metal amides such as lithium amide, sodium amide and potassium amide, alkali metal and alkaline earth metal carbonates such as lithium carbonate and calcium carbonate and also alkali metal hydrogen-30 carbonates such as sodium hydrogencarbonate, and organometallic compounds, in particular alkali metal alkyls such as methyllithium, butyllithium and phenyllithium, alkylmagnesium halides such as methylmagnesium chloride and also alkali metal and alkaline earth metal alkoxides 35 such as sodium methoxide, sodium ethoxide, potassium ethoxide, potassium tert-butoxide and dimethoxymagnesium, additionally organic bases, eg. tertiary amines such as trimethylamine, triethylamine, tri-isopropylethylamine and N-methylpiperidine, pyridine, substituted pyridines such as collidine, lutidine and 4-dimethylaminopyridine and also bicyclic amines.

Sodium hydrogencarbonate, sodium carbonate, triethylamine and pyridine are particularly preferred.

The bases are in general employed in equimolar amounts based on the compound II. However, they can also be used in an excess of from 5 mol% to 30 mol%, preferably 5 mol% to 10 mol%, or - in the case of the use of tertiary amines - if appropriate as a solvent.

The starting materials are in general reacted with one another in equimolar amounts. It may be advantageous for the yield to employ II in an excess of from 15 1 mol% to 20 mol%, preferably 1 mol% to 10 mol%, based on III.

The starting substances of the formulae II and III needed for preparing the compounds I are known in the literature (Houben-Weyl, Methoden der org. Chemie (Methods of Organic Chemistry), Vol. 10/1, pp. 1138-1148) or can be prepared according to the literature cited.

With respect to their use in fungicidal compositions, suitable compounds of the formula I are those in which the substituents have the following meanings:

is C2-C12-alkyl such as ethyl and straight-chain or 25 R branched propyl, butyl, pentyl, hexyl, heptyl, octyl, nonyl, decyl, undecyl and dodecyl, particularly straight-chain or branched  $C_3$ - $C_{10}$ -alkyl such as buty1, 1-methylpropyl, 1-methylethyl, propyl, n-pentyl, 1,1-dimethylethyl, 30 2-methylpropyl, 3-methylbutyl, 2-methylbutyl, 1-methylbutyl, 1,1-dimethylpropyl, 1,2-dimethylpropyl, n-hexyl, 1-ethylpropyl, 2,2-dimethylpropyl, 3-methylpentyl, 2-methylpentyl, 1-methylpentyl, 1,2-dimethylbutyl, 1,3-dimethyl-35 4-methylpentyl, 2,3-dimethylbutyl, 1,3-dimethylbutyl, 1,1-dimethylbutyl, 2,2-dimethylbutyl, 3,3-dimethyl- 5 - 0.Z. 0050/43543

butyl, 1,1,2-trimethylpropyl, 1,2,2-trimethylpropyl, 1-ethylbutyl, 2-ethylbutyl, 1-ethyl-3-methylpropyl, n-heptyl, 1-methylhexyl, 1-ethylpentyl, 2-ethylpentyl, 1-propylbutyl, octyl, 1-methylheptyl, 2-ethylhexyl, 2-methylheptyl, 1-ethylhexyl, 1-propylpentyl, 2-propylpentyl, nonyl, 1-methyloctyl, 2-methyloctyl, 1-ethylheptyl, 2-ethylheptyl, 1-propylhexyl, 2-propylhexyl, decyl, 1-methylnonyl, 2-methylnonyl, 1-ethyloctyl, 2-ethyloctyl, 1-propylheptyl and 2-propylheptyl, in particular propyl, 1-methylethyl, butyl, 1-methylbutyl, 2-methylbutyl, 1,1-dimethylethyl, pentyl, 1-methylbutyl, hexyl, heptyl and 1-methylheptyl, where these groups can be partially or completely halogenated, ie. hydrogens of these groups can be partially or completely replaced by halogens such as fluorine, chlorine and bromine, in particular fluorine and chlorine, for example haloalkyl such as chloromethyl, dichloromethyl, trichloromethyl, fluoromethyl, difluoromethyl, trifluoromethyl, chlorofluoromethyl, dichlorofluoromethyl, chlorodifluoromethyl, 1-fluoroethyl, 2-fluoroethyl, 2,2-difluoroethyl, 2,2,2-trifluoroethyl, 2-chloro-2-fluoroethyl, 2-chloro-2,2-difluoroethyl, 2,2-dichloro-2-fluoroethyl, 2,2,2-trichloroethyl and pentafluoroethyl;

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C2-C12-alkoxy such as ethoxy and straight-chain or branched propoxy, butoxy, pentoxy, hexyloxy, heptyloxy, octyloxy, nonyloxy, decyloxy, undecyloxy and dodecyloxy, particularly straight-chain or branched C2-C10-alkoxy such as ethoxy, propoxy, 1-methylethoxy, butoxy, 1-methylpropoxy, 2-methylpropoxy, 1-methylbutoxy, 1,1-dimethylethoxy, n-pentoxy, 2-methylbutoxy, 3-methylbutoxy, 1,2-dimethylpropoxy, 1-methylpentoxy, n-hexyloxy, 1-ethylpropoxy, 2-methylpentoxy, 3-methylpentoxy, 4-methylpentoxy, 1,3-dimethylbutoxy, 1,2-dimethylbutoxy,

1,2-dimethylbutoxy, 2,3-dimethylbutoxy, 2,2-dimethylbutoxy, 3,3-dimethylbutoxy, 1,1,2-tri-1,2,2-trimethylpropoxy, methylpropoxy, 1-ethyl-2-methylpropoxy, 2-ethylbutoxy, butoxy, 1-methylhexyloxy, 2-methylhexyloxy, n-heptyloxy, 3-methylhexyloxy, 4-methylhexyloxy, 5-methylhexyloxy, 1-ethylpentoxy, 2-ethylpentoxy, 1-propylbutoxy, 2-methylheptyloxy, 1-methylheptyloxy, octyloxy, 1-ethylhexyloxy, 2-ethylhexyloxy, 1-propylpentoxy, 1-methyloctyloxy, nonyloxy, 2-propylpentoxy, 2-methyloctyloxy, 1-ethylheptyloxy, 2-ethylheptyloxy, 1-propylhexyloxy, 2-propylhexyloxy, decyloxy, 1-methylnonyloxy, 2-methylnonyloxy, 1-ethyloctyloxy, 2-ethyloctyloxy, 1-propylheptyloxy and 2-propylheptyloxy, in particular ethoxy, propoxy, 1-methylethoxy, butoxy, 1-methylpropoxy, 2-methylpropoxy, 1,1-dimethylethoxy, pentoxy, hexyloxy and 2-ethylhexyloxy, where these groups can be partially or completely halogenated, ie. the hydrogens of these groups can be partially or completely replaced by halogens such as fluorine, chlorine and bromine, in particular fluorine and chlorine, for example halodichloromethoxy, alkoxy such as chloromethoxy, difluoromethoxy, trichloromethoxy, fluoromethoxy, dichlorochlorofluoromethoxy, trifluoromethoxy, 1-fluorochlorodifluoromethoxy, fluoromethoxy, 2,2-difluoroethoxy, 2-fluoroethoxy, ethoxy, 2-chloro-2-fluoroethoxy, 2,2,2-trifluoroethoxy, 2-chloro-2,2-difluoroethoxy, 2,2-dichloro-2-fluoroethoxy, 2,2,2-trichloroethoxy and pentafluoroethoxy;

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C<sub>1</sub>-C<sub>12</sub>-alkenyl such as straight-chain or branched propenyl, butenyl, pentenyl, hexenyl, heptenyl, octenyl, nonenyl, decenyl, undecenyl and dodecenyl, particularly straight-chain or branched C<sub>3</sub>-C<sub>10</sub>-alkenyl such as 2-propenyl, 2-butenyl, 3-butenyl, 1-methyl-2-propenyl, 2-methyl-2-propenyl,

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2-pentenyl,
                       3-pentenyl,
                                     4-pentenyl,
                                                   1-methyl-
         2-butenyl, 2-methyl-2-butenyl, 3-methyl-2-butenyl,
         1-methyl-3-butenyl, 2-methyl-3-butenyl,
                                                   3-methyl-
         3-butenyl, 1,1-dimethyl-2-propenyl, 1,2-dimethyl-
         2-propenyl,
                         1-ethyl-2-propenyl,
                                                  2-hexenyl,
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                                                   1-methyl-
         3-hexenyl,
                       4-hexenyl,
                                     5-hexenyl,
         2-pentenyl,
                         2-methyl-2-pentenyl,
                                                   3-methyl-
                                                   1-methyl-
         2-pentenyl,
                         4-methyl-2-pentenyl,
                                                   3-methyl-
         3-pentenyl,
                         2-methyl-3-pentenyl,
10
                         4-methyl-3-pentenyl,
                                                   1-methyl-
         3-pentenyl,
                                                   3-methyl-
         4-pentenyl,
                         2-methyl-4-pentenyl,
         4-pentenyl,
                        4-methyl-4-pentenyl,
                                               1,1-dimethyl-
                                               1,2-dimethyl-
                      1,1-dimethyl-3-butenyl,
         2-butenyl,
         2-buteny1,
                      1,2-dimethyl-3-butenyl,
                                               1,3-dimethyl-
                                               2,2-dimethyl-
15
                      1,3-dimethyl-3-butenyl,
         2-butenyl,
         3-butenyl,
                      2,3-dimethyl-2-butenyl,
                                              "2,3-dimethyl-
         3-butenyl,
                      1-ethyl-2-butenyl,
                                          1-ethyl-3-butenyl,
                              2-ethyl-3-butenyl,
                                                  1,1,2-tri-
         2-ethyl-2-butenyl,
                                1-ethyl-1-methyl-2-propenyl,
         methyl-2-propenyl,
                                        1-methyl-2-pentenyl,
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         1-ethyl-2-methyl-2-propenyl,
         2-methyl-2-pentenyl, 1-methyl-3-pentenyl, 2-methyl-
         3-pentenyl, 1-methyl-2-hexenyl, 2-methyl-2-hexenyl,
         1-methyl-3-hexenyl, 2-methyl-3-hexenyl, 1-ethyl-
         2-pentenyl, 2-ethyl-2-pentenyl, 1-ethyl-3-pentenyl,
         2-ethyl-3-pentenyl, 1-methyl-2-heptenyl, 2-methyl-
25
                         1-methyl-3-heptenyl,
                                                   2-methyl-
         2-heptenvl,
         3-heptenyl, 1-ethyl-2-hexenyl, 2-ethyl-2-hexenyl,
                              2-ethyl-3-hexenyl,
                                                   1-methyl-
         1-ethyl-3-hexenyl,
         2-octenyl, 2-methyl-2-octenyl, 1-methyl-3-octenyl,
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                               1-ethyl-2-heptenyl,
                                                    2-ethyl-
         2-methyl-3-octenyl,
         2-heptenyl, 1-ethyl-3-heptenyl, 2-ethyl-3-heptenyl,
                                                    1-ethyl-
                               2-ethyl-2-octenyl,
         1-ethyl-2-octenyl,
                                             in particular
         3-octenyl and 2-ethyl-3-octenyl,
         1-propenyl, 2-propenyl, 1-methylethenyl, 1-methyl-
         2-propenyl, 2-methyl-2-propenyl, 1-ethyl-2-propenyl,
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         1-methyl-2-butenyl, 1-ethyl-2-butenyl, 1-(1-methyl-
                                                   1-methyl-
                              1-buty1-2-buteny1,
         ethyl)-2-butenyl,
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2-pentenyl and 1,4-dimethyl-2-pentenyl, where these groups can be partially or completely halogenated, ie. the hydrogens of these groups can be partially or completely replaced by halogens such as fluorine, chlorine and bromine, in particular fluorine and chlorine, in particular 3-chloro-2-propenyl and 2,3-dichloro-2-propenyl;

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C3-C12-alkenyloxy such as straight-chain or branched propenyloxy, butenyloxy, pentenyloxy, hexenyloxy, decenyloxy, octenyloxy, nonenyloxy, heptenyloxy, 10 particularly and dodecenyloxy, undecenyloxy straight-chain or branched C3-C10-alkenyloxy such as 2-propenyloxy, 2-butenyloxy, 3-butenyloxy, 1-methyl-2-propenyloxy, 2-methyl-2-propenyloxy, 2-pentenyl-4-pentenyloxy, 1-methyl-3-pentenyloxy, 15 3-methyl-2-methyl-2-butenyloxy, 2-butenyloxy, 1-methyl-3-butenyloxy, 2-methyl-2-butenyloxy, 3-butenyloxy, 3-methyl-3-butenyloxy, 1,1-dimethyl-2-propenyloxy, 1,2-dimethyl-2-propenyloxy, 1-ethyl-2-hexenyloxy, 3-hexenyloxy, 20 2-propenyloxy, 4-hexenyloxy, 5-hexenyloxy, 1-methyl-2-pentenyloxy, 3-methyl-2-pentenyloxy, 2-methyl-2-pentenyloxy, 1-methyl-3-pentenyloxy, 4-methyl-2-pentenyloxy, 3-methyl-3-pentenyloxy, 2-methyl-3-pentenyloxy, 1-methyl-4-pentenyloxy, 4-methyl-3-pentenyloxy, 3-methyl-4-pentenyloxy, 2-methyl-4-pentenyloxy, 4-methyl-4-pentenyloxy, 1,1-dimethyl-2-butenyloxy, 1,2-dimethyl-1,1-dimethyl-3-butenyloxy, 2-butenyloxy, 1,2-dimethyl-3-butenyloxy, 1,3-dimethyl-2-butenyloxy, 1,3-dimethyl-3-butenyl-30 2,3-dimethyl-2,2-dimethyl-3-butenyloxy, оху, 2-butenyloxy, 2,3-dimethyl-3-butenyloxy, 1-ethyl-1-ethyl-3-butenyloxy, 2-butenyloxy, 2-butenyloxy, 2-ethyl-3-butenyloxy, 1,1,2-trimethyl-1-ethyl-1-methyl-2-propenyloxy, 2-propenyloxy, 35 1-ethy1-2-methyl-2-propenyloxy, 1-methyl-2-pentenyl-

oxy, 2-methyl-2-pentenyloxy, 1-methyl-3-pentenyloxy, 2-methyl-3-pentenyloxy, 1-methyl-2-hexenyloxy, 2-methy1-2-hexenyloxy, 1-methyl-3-hexenyloxy, 2-methyl-3-hexenyloxy, 1-ethyl-2-pentenyloxy, 5 2-ethyl-2-pentenyloxy, 1-ethyl-3-pentenyloxy, 2-ethyl-3-pentenyloxy, 1-methyl-2-heptenyloxy, 2-methy1-2-heptenyloxy, 1-methyl-3-heptenyloxy, 2-methyl-3-heptenyloxy, 1-ethyl-2-hexenyloxy, 2-ethyl-2-hexenyloxy, 1-ethyl-3-hexenyloxy, 2-ethyl-10 3-hexenyloxy, 1-methyl-2-octenyloxy, 2-methyl-2-octenyloxy, 1-methyl-3-octenyloxy, 2-methyl-3-octenyloxy, 1-ethyl-2-heptenyloxy, 2-ethyl-1-ethyl-3-heptenyloxy, 2-heptenyloxy, 2-ethyl-3-heptenyloxy, 1-ethyl-2-octenyloxy, 2-ethy1-15 1-ethyl-3-octenyloxy and 2-ethyl-2-octenyloxy, 3-octenyloxy, in particular 2-propenyloxy, 1-methyl-2-propenyloxy, 2-methyl-2-propenyloxy, 2-pentenyl-3-pentenyloxy, 1-methyl-2-butenyloxy 1-methyl-2-pentenyloxy, where these groups can be 20 partially or completely halogenated, hydrogens of these groups can be partially or completely replaced by halogens such as fluorine, chlorine and bromine, in particular fluorine and 3-chloro-2-propenyloxy, chlorine, in particular 2,3,3-trichloro-25 2,3-dichloro-2-propenyloxy and 2-propenyloxy;

C<sub>3</sub>-C<sub>6</sub>-alkynyl such as 2-propynyl, 2-butynyl, 3-butynyl, 1-methyl-2-propynyl, 2-pentynyl, 3-pentynyl, 4-pentynyl, 1-methyl-3-butynyl, 2-methyl-3-butynyl, 1-methyl-2-butynyl, 1,1-dimethyl-2-propynyl, 1-ethyl-2-propynyl, 2-hexynyl, 3-hexynyl, 4-alkynyl, 5-hexynyl, 1-methyl-2-pentynyl, 1-methyl-3-pentynyl, 1-methyl-4-pentynyl, 2-methyl-3-pentynyl, 2-methyl-4-pentynyl, 3-methyl-4-pentynyl, 4-methyl-2-pentynyl, 1,2-dimethyl-2-butynyl, 1,1-dimethyl-3-butynyl, 1,2-dimethyl-3-butynyl, 2,2-dimethyl-3-

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butynyl, 1-ethyl-2-butynyl, 1-ethyl-3-butynyl, 2-ethyl-3-butynyl and 1-ethyl-1-methyl-2-propynyl, in particular 2-propynyl, 2-butynyl and 3-butynyl, where these groups can be partially or completely halogenated, ie. the hydrogens of these groups can be partially or completely replaced by halogens such as fluorine, chlorine and bromine, in particular fluorine and chlorine, for example 3-chloro-2-propynyl, 3-chloro-2-butynyl and 4-chloro-3-butynyl;

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C3-C6-alkynyloxy such as 2-propynyloxy, 2-butynyloxy, 10 3-butynyloxy, 1-methyl-2-propynyloxy, 2-pentynyloxy, 3-pentynyloxy, 3-pentynyloxy, 4-pentynyloxy, 2-methyl-3-butynyloxy, methyl-3-butynyloxy, methyl-2-butynyloxy, 1,1-dimethyl-2-propionyloxy, 1ethyl-2-propynyloxy, 2-hexynyloxy, 3-hexynyloxy, 4-15 alkynyloxy, 5-hexynyloxy, 1-methyl-2-pentynyloxy, 1methyl-3-pentynyloxy, 1-methyl-4-pentynyloxy, 2-methyl-4-pentynyloxy, methyl-3-pentynyloxy, 4-methy1-3-pentynyloxy, 3-methyl-4-pentynyloxy, 1,1-dimethyl-2-butynyloxy, 1,1-dimethyl-3-butynyl-20 2,2-dimethyl-3-1,2-dimethyl-3-butynyloxy, оху, butynyloxy, 1-ethyl-2-butynyloxy, 1-ethyl-3-butynyloxy, 2-ethyl-3-butynyloxy and 1-ethyl-1-methyl-2propynyloxy, preferably 2-propynyloxy, 2-butynyloxy, 1-methyl-2-propynyloxy and 1-methyl-2-butynyloxy, 2-25 propynyloxy, 2-butynyloxy, 3-butynyloxy and methyl-2-propynyloxy, where these groups can be partially or completely halogenated, ie. the hydrogens of these groups can be partially or completely replaced by halogens such as fluorine, chlorine and 30 bromine, in particular fluorine and chlorine, for example 3-chloro-2-propynyloxy, 3-chloro-2-butynyloxy and 4-chloro-3-butynyloxy;

 $C_3$ - $C_7$ -cycloalkyl such as cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl and cycloheptyl, where these

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rings can carry one to 3 C<sub>1</sub>-C<sub>4</sub>-alkyls such as methyl, ethyl, propyl, 1-methylethyl, butyl, 1-methylpropyl, 2-methylpropyl and 1,1-dimethylethyl;

C<sub>4</sub>-C<sub>7</sub>-cycloalkenyl such as cyclobutenyl, cyclopentenyl, cyclohexenyl and cycloheptenyl, where these rings can carry one to three C<sub>1</sub>-C<sub>4</sub>-alkyls such as methyl, ethyl, propyl, 1-methylethyl, butyl, 1-methylpropyl, 2-methylpropyl and 1,1-dimethylethyl;

C<sub>3</sub>-C<sub>7</sub>-cycloalkoxy such as cyclopropoxy, cyclobutoxy, cyclopentoxy, cyclohexyloxy and cycloheptyloxy, where these rings can carry one to 3 C<sub>1</sub>-C<sub>4</sub>-alkyls such as methyl, ethyl, propyl, 1-methylethyl, butyl, 1-methylpropyl, 2-methylpropyl and 1,1-dimethylethyl;

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or C<sub>4</sub>-C<sub>7</sub>-cycloalkenyloxy such as 1-cyclobutenyloxy,
2-cyclobutenyloxy,
1-cyclopentenyloxy,
1-cyclopentenyloxy,
3-cyclopentenyloxy,
3-cyclohexenyloxy,
2-cyclohexenyloxy,
3-cycloheptenyloxy,
3-cycloheptenyloxy,
3-cycloheptenyloxy,
1-cycloheptenyloxy,
20 3-cycloheptenyloxy and 4-cycloheptenyloxy, where
these rings can carry one to 3 C<sub>1</sub>-C<sub>4</sub>-alkyls such as
methyl, ethyl, propyl, 1-methylethyl, butyl, 1methylpropyl, 2-methylpropyl and 1,1-dimethylethyl;

phenyl, which can carry one to five halogens such as fluorine, chlorine, bromine and iodine, in particular fluorine, chlorine and bromine, and/or one to three of the following radicals:

- C<sub>1</sub>-C<sub>4</sub>-alkyl as mentioned above;
- C<sub>1</sub>-C<sub>4</sub>-haloalkyl as mentioned above;
- 30 C<sub>1</sub>-C<sub>4</sub>-alkoxy as mentioned above;
  - C<sub>1</sub>-C<sub>4</sub>-haloalkoxy as mentioned above;
  - C1-C4-alkylthio such as methylthio, ethylthio,

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propylthio, 1-methylethylthio, butylthio, 1-methylpropylthio, 2-methylpropylthio and 1,1-dimethylethylthio;

- or C<sub>1</sub>-C<sub>4</sub>-haloalkylthio, particularly C<sub>1</sub>-C<sub>2</sub>-haloalkylthio such as chloromethylthio, dichloromethylthio, trichloromethylthio, fluoromethylthio, difluoromethylthio, trifluoromethylthio, chlorofluoromethylthio, dichlorofluoromethylthio, chlorodifluoromethylthio, 1-fluoroethylthio, 2-fluoroethylthio, 2,2-difluoroethylthio, 2,2,2-trifluoroethylthio, 2-chloro-2-fluoroethylthio, 2,2-dichloro-2-fluoroethylthio, 2,2-dichloro-2-fluoroethylthio, 2,2,2-trichloroethylthio and pentafluoroethylthio.
- 15 A is a cyclic radical from the group consisting of the formulae Al to A7:

where the substituents have the following meanings:

- X is -CH<sub>2</sub>-, -S-, -SO- or -SO<sub>2</sub>-;
- Y is -0- or -5-;

5

- 20 R<sup>1</sup>, R<sup>2</sup>, R<sup>4</sup>, R<sup>5</sup> and R<sup>7</sup> independently of one another are halogen such as fluorine, chlorine and bromine, C<sub>1</sub>-C<sub>4</sub>-alkyl as mentioned above, or C<sub>1</sub>-C<sub>4</sub>-haloalkyl as mentioned above;
- R<sup>3</sup> and R<sup>6</sup> independently of one another are hydrogen, 25 halogen such as fluorine, chlorine and bromine or

C1-C4-alkyl as mentioned above;

n is 1 or 2, where the radicals R<sup>3</sup> can be different if the value of n is 2.

- With respect to the biological action, particularly preferred compounds of the formula I are those in which R has the abovementioned meanings and A is a cyclic radical from the group consisting of the formulae A1 to A7, where X and Y have the abovementioned meaning and the substituents are the following radicals:
- 10 R¹ is halogen such as fluorine, chlorine and bromine, methyl or C₁-haloalkyl such as chloromethyl, dichloromethyl, trichloromethyl, fluoromethyl, difluoromethyl, trifluoromethyl, chlorofluoromethyl, dichlorofluoromethyl and chlorodifluoromethyl;
- is halogen such as fluorine, chlorine and bromine or C<sub>1</sub>-haloalkyl such as chloromethyl, dichloromethyl, trichloromethyl, fluoromethyl, difluoromethyl, trifluoromethyl, chlorofluoromethyl, dichlorofluoromethyl and chlorodifluoromethyl;
- 20 R3 is hydrogen or methyl;

35

- n is 1 or 2, where the radicals R<sup>3</sup> can be different if the value of n is 2;
- R<sup>4</sup> is halogen such as fluorine, chlorine and bromine or methyl;
- 25 R<sup>5</sup> is methyl or C<sub>1</sub>-haloalkyl such as chloromethyl, dichloromethyl, trichloromethyl, fluoromethyl, difluoromethyl, trifluoromethyl, chlorofluoromethyl, dichlorofluoromethyl and chlorodifluoromethyl;
- R<sup>6</sup> is hydrogen, halogen such as fluorine, chlorine and bromine or methyl;
  - R' is halogen such as fluorine, chlorine and bromine, methyl or C<sub>1</sub>-haloalkyl such as chloromethyl, dichloromethyl, trichloromethyl, fluoromethyl, difluoromethyl, trifluoromethyl, chlorofluoromethyl, dichlorofluoromethyl and chlorodifluoromethyl.

In particular, those compounds of the formula I are preferred in which R has the abovementioned meaning

and A is a cyclic radical from the group consisting of the formulae Al to A7, where X and Y have the abovementioned meaning and the substituents are the following groups:

- 5 R¹ is chlorine, bromine, iodine, methyl or trifluoromethyl;
  - R<sup>2</sup> is chlorine or trifluoromethyl;
  - R<sup>3</sup> is hydrogen or methyl;
  - n is 1 or 2, where the radicals R<sup>3</sup> can be different if the value of n is 2;
- the value of n is 2;
  R<sup>4</sup> is chlorine or methyl;
  - R<sup>5</sup> is methyl, difluoromethyl or trifluoromethyl;
  - R<sup>6</sup> is hydrogen, chlorine or methyl;
  - R' is chlorine, methyl or trifluoromethyl.
- Particularly preferred compounds of the formula I are summarized in the following Tables A to G.

Table A

5

1.1

10 ,		
]	R <sup>1</sup>	R
	CF <sub>3</sub>	i-C <sub>3</sub> H <sub>7</sub>
	CF <sub>3</sub>	n-C <sub>3</sub> H <sub>7</sub>
15	CF <sub>3</sub>	n-C <sub>4</sub> H <sub>9</sub>
	CF <sub>3</sub>	secC4H9
	CF <sub>3</sub>	i-C <sub>4</sub> H <sub>9</sub>
	CF <sub>3</sub>	tertC <sub>4</sub> H <sub>9</sub>
20	CF <sub>3</sub>	n-C <sub>5</sub> H <sub>11</sub>
	CF <sub>3</sub>	sec-C <sub>5</sub> H <sub>11</sub>
	CF3	n-C <sub>6</sub> H <sub>13</sub>
	CF3	n-C7H15
25	CF <sub>3</sub>	secC7H <sub>15</sub>
25	CF <sub>3</sub>	1-methylvinyl
	CF <sub>3</sub>	2-methylvinyl
	CF <sub>3</sub>	allyl
	CF <sub>3</sub>	2-methylallyl
30	CF <sub>3</sub>	2-ethylallyl
	CF <sub>3</sub>	1-methylallyl
. !	CF <sub>3</sub>	1-ethylallyl
	CF <sub>3</sub>	1-methyl-2-butenyl
35	CF3	1-ethy1-2-buteny1
	CF <sub>3</sub>	1-isopropyl-2-butenyl
	CF <sub>3</sub>	1-n-buty1-2-buteny1
	CF <sub>3</sub>	1-methyl-2-pentenyl
40	CF <sub>3</sub>	1,4-dimethyl-2-pentenyl
	CF <sub>3</sub>	propargyl
	CF <sub>3</sub>	2-butyny1
	CF <sub>3</sub>	3-butyny1
	CF <sub>3</sub>	ethoxy
45	CF <sub>3</sub>	ргорожу
	CF <sub>3</sub>	1-methylethoxy

1		<del> </del>
	R <sup>1</sup>	R
	CF <sub>3</sub>	n-butoxy
_	CF <sub>3</sub>	1-methylpropoxy
5	CF <sub>3</sub>	2-methylpropoxy
	CF <sub>3</sub>	1,1-dimethylethoxy
	CF <sub>3</sub>	n-pentyloxy
	CF <sub>3</sub>	n-hexyloxy
10	CF <sub>3</sub>	2-ethylhexyloxy
	CF <sub>3</sub>	2-propenyloxy
	CF <sub>3</sub>	2-butenyloxy
	CF <sub>3</sub>	2-methyl-2-propenyloxy
15	CF3	2-pentenyloxy
	CF <sub>3</sub>	3-pentenyloxy
	CF3	3-chloro-2-propenyloxy
	CF <sub>3</sub>	2,3-dichloro-2-propenyloxy
20	CF <sub>3</sub>	2,3,3-trichloropropenyloxy
	CF3	2-propynyloxy
	CF <sub>3</sub>	2-butynyloxy
	CF <sub>3</sub>	3-butynyloxy
	CF <sub>3</sub>	1-methy1-2-propynyloxy
25	CF <sub>3</sub>	cyclopropyl
	CF3	cyclobutyl
	CF <sub>3</sub>	cyclopentyl
	CF <sub>3</sub>	cyclohexyl
30	CF <sub>3</sub>	2-cyclopentenyl
	CF <sub>3</sub>	1-cyclopentenyl
	CF <sub>3</sub>	2-cyclohexenyl
	CF3	1-cyclohexenyl
35	CF <sub>3</sub>	cyclopentyloxy
	CF <sub>3</sub>	cyclohexyloxy
	CF <sub>3</sub>	2-cyclopentenyloxy
	CF <sub>3</sub>	2-cyclohexenyloxy
40	CF <sub>3</sub>	phenyl
40	C1 ·	i-C <sub>3</sub> H <sub>7</sub>
	C1	n-C <sub>3</sub> H <sub>7</sub>
	C1	n-C <sub>4</sub> H <sub>9</sub>
	Cl	secC <sub>4</sub> H <sub>9</sub>
45	Cl	i-C <sub>4</sub> H <sub>9</sub>
	Cl	tertC4H9
		•

ı	R <sup>1</sup>	
	<b>K</b> -	R
	Cl	n-C <sub>5</sub> H <sub>11</sub>
۰	Cl	secC <sub>5</sub> H <sub>11</sub>
5	C1	n-C <sub>6</sub> H <sub>13</sub>
	C1	n-C <sub>7</sub> H <sub>15</sub>
	C1	secC7H15
- {	C1	1-methylvinyl
10	C1	2-methylvinyl
	C1	allyl
	Cl	2-methylvinyl
ļ	C1	2-ethylallyl
15	Cl	1-methylallyl
	C1	1-ethylallyl
	Cl	1-methyl-2-butenyl
	C1	1-ethyl-2-butenyl
20	Cl	1-isopropy1-2-buteny1
	C1	1-n-buty1-2-buteny1
	C1	methyl-2-pentenyl
	Cl	1,4-dimethyl-2-pentenyl
	Cl	propargyl
25	Cl	2-butyny1
- {	Cl	3-butyny1
	Cl	ethoxy
	C1	propoxy
30	C1	1-methylethoxy
	Cl	n-butoxy
	C1	1-methylpropoxy
	Cl	2-methylpropoxy
35	C1	1,1-dimethylethoxy
	C1	n-pentyloxy
	C1	n-hexyloxy
	C1	2-ethylhexyloxy
40	C1	2-propenyloxy
40	C1	2-butenyloxy
1	Cl	2-methy1-2-propenyloxy
l	Cl	2-pentenyloxy
	Cl	3-pentenyloxy
45	Cl	3-chloro-2-propenyloxy
	C1	2,3-dichloro-2-propenyloxy

	R <sup>1</sup>	R
	Cl	2,3,3-trichloropropenyloxy
_ [	Cl	2-propynyloxy
5	Cl	2-butynyloxy
	Cl	3-butynyloxy
	Cl	1-methy1-2-propynyloxy
Γ	C1	cyclopropyl
10	Cl	cyclobutyl
Γ	Cl	cyclopentyl
	Cl	cyclohexyl
Γ	Cl	2-cyclopentenyl
15	C1	1-cyclopenteny1
Γ	Cl	2-cyclohexenyl
Γ	Cl	1-cyclohexenyl
	Cl	cyclopentyloxy
20	Cl	cyclohexyloxy
<b>~</b> [	C1	2-cyclopentenyloxy
Γ	Cl	2-cyclohexenyloxy
Γ	Cl	phenyl

and the second s

A E

Table B

10	R <sup>2</sup>	R
	C1	i-C <sub>3</sub> H <sub>7</sub>
	C1	n-C <sub>3</sub> H <sub>7</sub>
	Cl	n-C4H9
15	Cl	secC <sub>4</sub> H <sub>9</sub>
	Cl	i-C <sub>4</sub> H <sub>9</sub>
	Cl	tertC <sub>4</sub> H <sub>9</sub>
	C1	n-C <sub>5</sub> H <sub>11</sub>
20	C1	secC <sub>5</sub> H <sub>11</sub>
	Cl	n-C <sub>6</sub> H <sub>13</sub>
	Cl	n-C <sub>7</sub> H <sub>15</sub>
	. C1	secC <sub>7</sub> H <sub>15</sub>
25	C1	1-methylvinyl
	C1	2-methylvinyl
	C1	allyl
	C1	2-methylallyl
	C1	2-ethylallyl
3.0	C1	1-methylallyl
	Cl	1-ethylallyl
	Cl	1-methy1-2-buteny1
	C1	1-ethyl-2-butenyl
35		1-isopropyl-2-butenyl
l	Cl	1-n-buty1-2-buteny1
		1-methyl-2-pentenyl
	C1	1,4-dimethyl-2-pentenyl
40		propargyl
- 1		2-butyny1
L		3-butyny1
1		ethoxy
45		ргороху
[		1-methylethoxy
L	C1	n-butoxy

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		40
	R <sup>2</sup>	R
	Cl	1-methylpropoxy
_	Cl	2-methylpropoxy
5	C1	1,1-dimethylethoxy
	Cl	n-pentyloxy
	Cl	n-hexyloxy
	Cl	2-ethylhexyloxy
10	· C1	2-propenyloxy
	Cl	2-butenyloxy
	Cl	2-methy1-2-propenyloxy
	Cl	2-pentenyloxy
15	Cl	3-pentenyloxy
	Cl	3-chloro-2-propenyloxy
	Cl	2,3-dichloro-2-propenyloxy
	CŢ	2,3,3-trichloropropenyloxy
20	Cl	2-propynyloxy
	Cl	2-butynyloxy
	Cl	3-butynyloxy
	C1	1-methyl-2-propynyloxy
	Cl	cyclopropyl
25	C1	cyclobutyl
	C1	cyclopentyl
	.Cl	cyclohexyl
	Cl	2-cyclopentenyl
30	C1	1-cyclopenteny1
	Cl	2-cyclohexenyl
	Cl	1-cyclohexenyl
	C1	cyclopentyloxy
35	C1	cyclohexyloxy
	C1	2-cyclopentenyloxy
	Cl	2-cyclohexenyloxy
	Cl	i-C <sub>3</sub> H <sub>7</sub>
40	Cl	n-C <sub>3</sub> H <sub>7</sub>
40	C1	n-C4H9
	Cl	secC4H9
	Cl .	i-C <sub>4</sub> H <sub>9</sub>
	C1	tertC <sub>4</sub> H <sub>9</sub>
45	C1	n-C <sub>5</sub> H <sub>11</sub>
	Cl	secC <sub>5</sub> H <sub>11</sub>

	R <sup>2</sup>	R
	Cl	n-C <sub>6</sub> H <sub>13</sub>
5	Cl	n-C <sub>7</sub> H <sub>15</sub>
3	C1	secC <sub>7</sub> H <sub>15</sub>
	C1	ethoxy
	Cl	propoxy
	C1	1-methylethoxy
10	C1	n-butoxy
	C1	1-methylpropoxy
	Cl	2-methylpropoxy
	C1	1,1-dimethylethoxy
15	C1	n-pentyloxy
	C1	n-hexyloxy
	C1	cyclopentyl
	C1	phenyl

Table C

5 
$$R \longrightarrow X \longrightarrow 0$$
 I.3

10 Х CH<sub>2</sub> i-C<sub>3</sub>H<sub>7</sub> CH<sub>2</sub> n-C3H7 15 CH<sub>2</sub> n-C4H9 CH<sub>2</sub> sec.-C4H9 CH<sub>2</sub> i-C<sub>4</sub>H<sub>9</sub> tert.-C<sub>4</sub>H<sub>9</sub> CH<sub>2</sub> 20 CH<sub>2</sub> n-C5H11 CH<sub>2</sub>  $sec.-C_5H_{11}$ CH<sub>2</sub> n-C<sub>6</sub>H<sub>13</sub> CH<sub>2</sub>  $n-C_7H_{15}$ CH<sub>2</sub> sec.-C7H15 CH<sub>2</sub> 1-methylvinyl CH<sub>2</sub> 2-methylvinyl CH<sub>2</sub> allyl 2-methylallyl CH<sub>2</sub> 30 CH2. 2-ethylallyl CH<sub>2</sub> 1-methylallyl CH2 1-ethylallyl CH<sub>2</sub> 1-methy1-2-buteny1 CH<sub>2</sub> 1-ethyl-2-butenyl CH<sub>2</sub> 1-isopropy1-2-buteny1 CH<sub>2</sub> 1-n-butyl-2-butenyl CH<sub>2</sub> 1-methy1-2-penteny1 CH<sub>2</sub> 1,4-dimethy1-2-penteny1 CH<sub>2</sub> propargyl CH<sub>2</sub> 2-butyny1: CH<sub>2</sub> 3-butyny1 CH<sub>2</sub> ethoxy CH<sub>2</sub> propoxy

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		23		
	х	R		
	CH <sub>2</sub>	1-methylethoxy		
5	CH <sub>2</sub>	n-butoxy		
J	CH <sub>2</sub>	1-methylpropoxy		
	CH <sub>2</sub>	2-methylpropoxy		
	CH <sub>2</sub>	1,1-dimethylethoxy		
	CH <sub>2</sub>	n-pentyloxy		
10	CH <sub>2</sub>	n-hexyloxy		
	CH <sub>2</sub>	2-ethylhexyloxy		
	CH <sub>2</sub>	2-propenyloxy		
	CH <sub>2</sub>	2-butenyloxy		
15	СH <sub>2</sub>	2-methy1-2-propenyloxy		
	CH <sub>2</sub>	2-pentenyloxy		
	CH <sub>2</sub>	3-pentenyloxy		
	CH <sub>2</sub>	3-chloro-2-propenyloxy		
20	CH <sub>2</sub>	2,3-dichloro-2-propenyloxy		
	CH <sub>2</sub>	2,3,3-trichloropropenyloxy		
	CH <sub>2</sub>	2-propynyloxy		
	CH <sub>2</sub>	2-butynyloxy		
25	CH <sub>2</sub>	3-butynyloxy		
23	CH <sub>2</sub>	1-methy1-2-propynyloxy		
	CH <sub>2</sub>	cyclopropyl		
	CH <sub>2</sub>	cyclobutyl		
	CH <sub>2</sub>	cyclopentyl		
30	CH <sub>2</sub>	cyclohexyl		
	CH <sub>2</sub>	2-cyclopentenyl		
	CH <sub>2</sub>	1-cyclopentenyl		
	CH <sub>2</sub>	2-cyclohexenyl		
35		1-cyclohexenyl		
L	CH <sub>2</sub>	cyclopentyloxy		
L		cyclohexyloxy		
Ĺ	CH <sub>2</sub>	2-cyclopentenyloxy		
40	CH <sub>2</sub>	2-cyclohexenyloxy		
	S	i-C <sub>3</sub> H <sub>7</sub>		
	S	n-C <sub>3</sub> H <sub>7</sub>		
_	S	n-C4H9		
<u>,                                    </u>	S s	BecC <sub>4</sub> H <sub>9</sub>		
45		L-C <sub>4</sub> H <sub>9</sub>		
L	S t	certC <sub>4</sub> H <sub>9</sub>		

	Х	R
	S	n-C <sub>5</sub> H <sub>11</sub>
5	S	secC <sub>5</sub> H <sub>11</sub>
3	S	n-C <sub>6</sub> H <sub>13</sub>
	S	n-C <sub>7</sub> H <sub>15</sub>
	S	secC7H <sub>15</sub>
	S	ethoxy
10	S	ргороху
	S	1-methylethoxy
	S	n-butoxy
	S	1-methylpropoxy
15	S	2-methylpropoxy
	S	1,1-dimethylethoxy
	S	n-pentyloxy
	S	n-hexyloxy
20	S	cyclopenty1
	S	phenyl

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Table D

	. R <sup>3</sup>	R <sup>3</sup>	
	Н	i-C <sub>3</sub> H <sub>7</sub>	0
15	Н	n-C <sub>3</sub> H <sub>7</sub>	0
	Н	n-C <sub>4</sub> H <sub>9</sub>	0
	Н	secC <sub>4</sub> H <sub>9</sub>	0
	Н	i-C <sub>4</sub> H <sub>9</sub>	0
20	Н	tertC <sub>4</sub> H <sub>9</sub>	0
-•	Н	n-C <sub>5</sub> H <sub>11</sub>	0
	Н	secC <sub>5</sub> H <sub>11</sub>	0
	Н	n-C <sub>6</sub> H <sub>13</sub>	0 .
	Н	n-C7H15	0
25	H	secC <sub>7</sub> H <sub>15</sub>	0
	Н	ethoxy	. 0 .
	H	propoxy	0
	Н	1-methylethoxy	. 0
30	Н	n-butoxy	0
	н	1-methylpropoxy	0
	Н	2-methylpropoxy	0
		1,1-dimethylethoxy	0
35	н	n-pentyloxy	. 0
	Н	n-hexyloxy	0
	Н	cyclopentyl	0
	H	cyclohexyl	0
[	Н	2-cyclopentenyl	
40	Н	1-cyclopentenyl	
, [	Н	2-cyclohexenyl O	
	Н	1-cyclohexenyl 0	
	H	cyclopentyloxy	
15 [	H	cyclohexyloxy	0
	Н	2-cyclopentenyloxy	0

	R <sup>3</sup>	R	Y
		~	1
	н	2-cyclohexenyloxy	0
5	CH <sub>3</sub>	i-C <sub>3</sub> H <sub>7</sub>	0
3	CH <sub>3</sub>	n-C <sub>3</sub> H <sub>7</sub>	0
	CH <sub>3</sub>	n-C <sub>4</sub> H <sub>9</sub>	0
	CH <sub>3</sub>	secC4H9	0
	CH <sub>3</sub>	i-C <sub>4</sub> H <sub>9</sub>	0
10	CH <sub>3</sub>	tertC <sub>4</sub> H <sub>9</sub>	0
	CH <sub>3</sub>	n-C <sub>5</sub> H <sub>11</sub>	0
	CH <sub>3</sub>	secC <sub>5</sub> H <sub>11</sub>	0
į	CH <sub>3</sub>	n-C <sub>6</sub> H <sub>13</sub>	0
15	CH <sub>3</sub>	n-C7H <sub>15</sub>	0
i	CH <sub>3</sub>	secC7H <sub>15</sub>	0
	CH <sub>3</sub>	ethoxy	0
	CH <sub>3</sub>	propoxy	0
20	CH <sub>3</sub>	1-methylethoxy	0
	CH <sub>3</sub>	n-butoxy	0
	CH <sub>3</sub>	1-methylpropoxy	0
j	CH <sub>3</sub>	2-methylpropoxy	0
25	CH <sub>3</sub>	1,1-dimethylethoxy	0
	CH <sub>3</sub>	n-pentyloxy	0
	CH <sub>3</sub>	n-hexyloxy	0
	CH <sub>3</sub>	cyclopentyl	0
	CH <sub>3</sub>	phenyl	0

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Table E

R<sup>4</sup>
HO N CO

I.5

10	R <sup>4</sup>	R	Y	
	CH <sub>3</sub>	i-C <sub>3</sub> H <sub>7</sub>	0	
	CH <sub>3</sub>	n-C <sub>3</sub> H <sub>7</sub>	0	
	CH <sub>3</sub>	n-C <sub>4</sub> H <sub>9</sub>	0	
15	CH <sub>3</sub>	secC <sub>4</sub> H <sub>9</sub>	0	
}	CH <sub>3</sub>	i-C <sub>4</sub> H <sub>9</sub>	0	
	CH <sub>3</sub>	tertC <sub>4</sub> H <sub>9</sub>	0	
	CH <sub>3</sub>	n-C <sub>5</sub> H <sub>11</sub>	0	
20	CH <sub>3</sub>	secC <sub>5</sub> H <sub>11</sub>	0	
ļ	CH <sub>3</sub>	n-C <sub>6</sub> H <sub>13</sub>	0	
	CH <sub>3</sub>	n-C <sub>7</sub> H <sub>15</sub>	0	
	CH <sub>3</sub>	secC7H15	0	
25	CH <sub>3</sub>	ethoxy	0	
	CH <sub>3</sub>	propoxy	0	
ĺ	CH <sub>3</sub>	1-methylethoxy	0	
	CH <sub>3</sub>	n-butoxy	0	
[	CH <sub>3</sub>	1-methylpropoxy	0	
30	CH <sub>3</sub>	2-methylpropoxy	0	
• [	CH <sub>3</sub>	1,1-dimethylethoxy	0	
	CH <sub>3</sub>	n-pentyloxy	0	
	CH <sub>3</sub>	n-hexyloxy		
35	CH₃	cyclopentyl		
	CH₃	cyclopentenyl	0	
[	CH <sub>3</sub>	i-C <sub>3</sub> H <sub>7</sub>		
	CH <sub>3</sub>	n-C <sub>3</sub> H <sub>7</sub> S		
40	CH <sub>3</sub>			
	CH <sub>3</sub>	secC <sub>4</sub> H <sub>9</sub>	S S	
	CH <sub>3</sub>	i-C <sub>4</sub> H <sub>9</sub>	s	
	CH <sub>3</sub>	tertC <sub>4</sub> H <sub>9</sub>	s	
٦. ٢	CH <sub>3</sub>	n-C <sub>5</sub> H <sub>11</sub>	s	
45	CH <sub>3</sub>	secC <sub>5</sub> H <sub>11</sub>	$\frac{s}{s}$	
Γ	CH <sub>3</sub>	n-C <sub>6</sub> H <sub>13</sub>	s	

	R <sup>4</sup>	R	Y
	CH <sub>3</sub>	n-C <sub>7</sub> H <sub>15</sub>	S
_	CH <sub>3</sub>	secC7H15	S
5	CH <sub>3</sub>	ethoxy	S
	CH <sub>3</sub>	propoxy	S
	CH₃	1-methylethoxy	S
	CH <sub>3</sub>	n-butoxy	S
10	СН3	1-methylpropoxy	S
	CH <sub>3</sub>	2-methylpropoxy	S
	CH₃	1,1-dimethylethoxy	S
15	CH <sub>3</sub>	n-pentyloxy	s
	CH₃	n-hexyloxy	·S
	CH <sub>3</sub>	cyclopentyl	S
	CH₃	cyclopentenyl	S

Table F

 $R^{6}$  N  $CH_{3}$  I.6

	R <sup>5</sup>	R <sup>6</sup>	R
15	CH <sub>3</sub>	н	i-C <sub>3</sub> H <sub>7</sub>
	CH <sub>3</sub>	н	n-C <sub>3</sub> H <sub>7</sub>
	CH <sub>3</sub>	Н	n-C <sub>4</sub> H <sub>9</sub>
	CH <sub>3</sub>	Н	secC <sub>4</sub> H <sub>9</sub>
	CH <sub>3</sub>	Н	1-C <sub>4</sub> H <sub>9</sub>
20	CH <sub>3</sub>	н	tertC4H9
	CH <sub>3</sub>	Н	n-C <sub>5</sub> H <sub>11</sub>
	CH <sub>3</sub>	Н	secC <sub>5</sub> H <sub>11</sub>
	CH <sub>3</sub>	Н	n-C <sub>6</sub> H <sub>13</sub>
25	СН3	Н	n-C7H15
	CH <sub>3</sub>	Н	secC <sub>7</sub> H <sub>15</sub>
	CH <sub>3</sub>	Н	1-methylvinyl
	CH <sub>3</sub>	Н	2-methylvinyl
L	CH <sub>3</sub>	Н	allyl
30	CH <sub>3</sub>	Н	2-methylallyl
L	CH <sub>3</sub>	Н	2-ethylallyl
	СН3	H	1-methylallyl
	CH <sub>3</sub>	Н	1-ethylallyl
35 L	CH <sub>3</sub>	Н	1-methyl-2-butenyl
L	CH <sub>3</sub>	Н	1-ethyl-2-butenyl
	CH <sub>3</sub>	Н	1-isopropyl-2-butenyl
	CH <sub>3</sub>	Н	1-n-buty1-2-buteny1
40	CH <sub>3</sub>	Н	1-methyl-2-pentenyl
**	CH <sub>3</sub>	Н	1,4-dimethy1-2-penteny1
L	CH <sub>3</sub>	Н	propargyl
	CH <sub>3</sub>	Н	2-butyny1
	CH <sub>3</sub>	Н	3-butynyl
45	CH <sub>3</sub>	н	ethoxy
	CH <sub>3</sub>	Н	ргороху

<sub>30</sub> 2106497

	R <sup>5</sup>	R <sup>6</sup>	30
	R3	К°	R
	CH <sub>3</sub>	Н	1-methylethoxy
5	CH <sub>3</sub>	Н	n-butoxy
•	CH <sub>3</sub>	Н	1-methylpropoxy
	CH₃	Н	2-methylpropoxy
	CH <sub>3</sub>	Н	1,1-dimethylethoxy
	CH <sub>3</sub>	Н	n-pentyloxy
10	CH <sub>3</sub>	н	n-hexyloxy
	CH <sub>3</sub>	н	2-ethylhexyloxy
	CH <sub>3</sub>	Н	2-propenyloxy
	CH <sub>3</sub>	н	2-butenyloxy
15	CH <sub>3</sub>	Н	2-methyl-2-propenyloxy
	CH <sub>3</sub>	н	2-pentenyloxy
	CH <sub>3</sub>	H	3-pentenyloxy
	CH <sub>3</sub>	н	3-chloro-2-propenyloxy
20	CH <sub>3</sub>	Н	2,3-dichloro-2-propenyloxy
	CH <sub>3</sub>	Н	2,3,3-trichloropropenyloxy
.	CH <sub>3</sub>	Н	2-propynyloxy
	CH <sub>3</sub>	н	2-butynyloxy
	CH <sub>3</sub>	Н	3-butynyloxy
25	CH <sub>3</sub>	H	1-methyl-2-propynyloxy
	CH <sub>3</sub>	H	cyclopropyl
	CH <sub>3</sub>	H	cyclobutyl
	CH <sub>3</sub>	Н	cyclopentyl
30	CH <sub>3</sub>	Н	cyclohexyl
	CH <sub>3</sub>	Н	2-cyclopentenyl
	CH <sub>3</sub>	H	1-cyclopentenyl
Ĺ	CH <sub>3</sub>	н	2-cyclohexenyl
35	CH <sub>3</sub>	Н	1-cyclohexenyl
	CH <sub>3</sub>	H	cyclopentyloxy
	CH <sub>3.</sub>	н	cyclohexyloxy
	CH <sub>3</sub>	Н	2-cyclopentenyloxy
40	CH <sub>3</sub>	н	2-cyclohexenyloxy
•	CF <sub>3</sub>	Н	i-C <sub>3</sub> H <sub>7</sub>
	CF <sub>3</sub>	Н	n-C <sub>3</sub> H <sub>7</sub>
	CF <sub>3</sub>	н	n-C <sub>4</sub> H <sub>9</sub>
ĺ	CF <sub>3</sub>	Н	secC <sub>4</sub> H <sub>9</sub>
45	CF <sub>3</sub>	Н	i-C <sub>4</sub> H <sub>9</sub>
ſ	CF <sub>3</sub>	Н	tertC <sub>4</sub> H <sub>9</sub>
			<del></del>

		21 MIGGIO.
R <sup>5</sup>	R <sup>6</sup>	R
CF <sub>3</sub>	н	n-C <sub>5</sub> H <sub>11</sub>
CF <sub>3</sub>	Н	secC <sub>5</sub> H <sub>11</sub>
CF <sub>3</sub>	Н	n-C <sub>6</sub> H <sub>13</sub>
CF <sub>3</sub>	Н	n-C <sub>7</sub> H <sub>15</sub>
CF <sub>3</sub>	Н	secC7H15
CF <sub>3</sub>	Н	ethoxy
CF <sub>3</sub>	Н	ргороху
	H	1-methylethoxy
CF3	Н	n-butoxy
CF <sub>3</sub>	Н	1-methylpropoxy
CF <sub>3</sub>	Н	2-methylpropoxy
CF <sub>3</sub>	Н	1,1-dimethylethoxy
	Н	n-pentyloxy
	Н	n-hexyloxy
The second secon	Н	cyclopentyl
	Н	cyclopentenyl
CF <sub>3</sub>	Н	pheny1
	CF3	CF3 H

Table G

5 
$$R^7$$
  $N$   $R^6$  1.7

10	R <sup>7</sup>	R6	R		
	CF <sub>3</sub>	CH <sub>3</sub>	i-C <sub>3</sub> H <sub>7</sub>		
	CF <sub>3</sub>	СН₃	n-C <sub>3</sub> H <sub>7</sub>		
15	CF <sub>3</sub>	CH <sub>3</sub>	n-C <sub>4</sub> H <sub>9</sub>		
	CF <sub>3</sub>	СН3	secC4H9		
	CF <sub>3</sub>	CH <sub>3</sub>	i-C <sub>4</sub> H <sub>9</sub>		
	CF <sub>3</sub>	CH <sub>3</sub>	tertC4H9		
20	CF <sub>3</sub>	CH <sub>3</sub>	n-C <sub>5</sub> H <sub>11</sub>		
	CF <sub>3</sub>	СН3	secC5H11		
[	CF <sub>3</sub>	CH <sub>3</sub>	n-C <sub>6</sub> H <sub>13</sub>		
. [	CF <sub>3</sub>	СН3	n-C7H15		
	CF <sub>3</sub>	CH <sub>3</sub>	secC7H <sub>15</sub>		
25	CF <sub>3</sub>	CH <sub>3</sub>	1-methylvinyl		
	CF <sub>3</sub>	CH <sub>3</sub>	2-methylvinyl		
	CF <sub>3</sub>	CH <sub>3</sub>	allyl		
	CF <sub>3</sub>	CH <sub>3</sub>	2-methylallyl		
30	CF <sub>3</sub>	CH <sub>3</sub>	2-ethylallyl		
	CF <sub>3</sub>	CH <sub>3</sub>	1-methylallyl		
	CF <sub>3</sub>	CH <sub>3</sub>	1-ethylallyl		
: · [	CF <sub>3</sub>	CH <sub>3</sub>	1-methyl-2-butenyl		
35	CF <sub>3</sub>	CH <sub>3</sub>	1-ethyl-2-butenyl		
. [	CF <sub>3</sub>	CH <sub>3</sub>	1-isopropyl-2-butenyl		
- [	CF <sub>3</sub>	CH <sub>3</sub>	1-n-buty1-2-buteny1		
: 4	CF <sub>3</sub>	СН3	1-methy1-2-penteny1		
40	CF <sub>3</sub>	CH <sub>3</sub>	1,4-dimethyl-2-pentenyl		
•	CF <sub>3</sub>	CH <sub>3</sub>	propargyl		
. [	CF <sub>3</sub>	CH <sub>3</sub>	2-butynyl		
	CF <sub>3</sub>	CH <sub>3</sub>	3-butynyl		
	CF <sub>3</sub>	CH <sub>3</sub>	ethoxy		
45	CF <sub>3</sub>	CH <sub>3</sub>	propoxy		
Ī	CF <sub>3</sub>	CH <sub>3</sub>	1-methylethoxy		

	R <sup>7</sup>	R <sup>6</sup>	R		
5	CF <sub>3</sub>	CH <sub>3</sub>	n-butoxy		
	CF <sub>3</sub>	CH <sub>3</sub>	1-methylpropoxy		
	CF <sub>3</sub>	CH <sub>3</sub>	2-methylpropoxy		
	CF <sub>3</sub>	CH <sub>3</sub>	1,1-dimethylethoxy		
	CF <sub>3</sub>	CH <sub>3</sub>	n-pentyloxy		
10	CF <sub>3</sub>	CH <sub>3</sub>	n-hexyloxy		
	CF₃	CH <sub>3</sub>	2-ethylhexyloxy		
	CF <sub>3</sub>	CH <sub>3</sub>	2-propenyloxy		
	CF <sub>3</sub>	CH <sub>3</sub>	2-butenyloxy		
	CF <sub>3</sub>	CH <sub>3</sub>	2-methy1-2-propenyloxy		
15	CF <sub>3</sub>	CH <sub>3</sub>	2-pentenyloxy		
	CF <sub>3</sub>	CH <sub>3</sub>	3-pentenyloxy		
ļ	CF <sub>3</sub>	CH <sub>3</sub>	3-chloro-2-propenyloxy		
į	CF <sub>3</sub>	CH <sub>3</sub>	2,3-dichloro-2-propenyloxy		
20	CF <sub>3</sub>	CH <sub>3</sub>	2,3,3-trichloropropenyloxy		
	CF <sub>3</sub>	CH <sub>3</sub>	2-propynyloxy		
	CF <sub>3</sub>	CH <sub>3</sub>	2-butynyloxy		
	CF <sub>3</sub>	CH <sub>3</sub>	3-butynyloxy		
25	CF <sub>3</sub>	CH <sub>3</sub>	1-methyl-2-propynyloxy		
	CF <sub>3</sub>	CH <sub>3</sub>	cyclopropyl		
L	CF <sub>3</sub>	CH <sub>3</sub>	cyclobutyl		
L	CF <sub>3</sub>	CH <sub>3</sub>	cyclopentyl		
L	CF <sub>3</sub>	CH <sub>3</sub>	cyclohexyl		
30	CF <sub>3</sub>	CH <sub>3</sub>	2-cyclopentenyl		
L	CF <sub>3</sub>	CH <sub>3</sub>	1-cyclopentenyl		
L	CF <sub>3</sub>	CH <sub>3</sub>	2-cyclohexenyl		
L	CF <sub>3</sub>	CH <sub>3</sub>	1-cyclohexenyl		
35	CF <sub>3</sub>	CH <sub>3</sub>	cyclopentyloxy		
	CF <sub>3</sub>	CH <sub>3</sub>	cyclohexyloxy		
	CF <sub>3</sub>	CH <sub>3</sub>	2-cyclopentenyloxy		
L	CF <sub>3</sub>	CH <sub>3</sub>	2-cyclohexenyloxy		
40	CH <sub>3</sub>	СН₃	i-C <sub>3</sub> H <sub>7</sub>		
	CH <sub>3</sub>	CH <sub>3</sub>	n-C <sub>3</sub> H <sub>7</sub>		
	CH <sub>3</sub>	CH <sub>3</sub>	n-C <sub>4</sub> H <sub>9</sub>		
$\Gamma$	CH <sub>3</sub>	CH <sub>3</sub>	secC <sub>4</sub> H <sub>9</sub>		
	CH <sub>3</sub>	СН3	i-C <sub>4</sub> H <sub>9</sub>		
45	CH <sub>3</sub>	CH <sub>3</sub>	tertC <sub>4</sub> H <sub>9</sub>		
Γ	CH <sub>3</sub>	CH <sub>3</sub>	n-C <sub>5</sub> H <sub>11</sub>		
-					

	R <sup>7</sup>	R <sup>6</sup>	R	
	CH <sub>3</sub>	CH <sub>3</sub>	secC <sub>5</sub> H <sub>11</sub>	
5	CH <sub>3</sub>	CH <sub>3</sub>	n-C <sub>6</sub> H <sub>13</sub>	
	CH <sub>3</sub>	CH <sub>3</sub>	n-C <sub>7</sub> H <sub>15</sub>	
	CH <sub>3</sub>	CH <sub>3</sub>	secC7H <sub>15</sub>	
	CH <sub>3</sub>	CH <sub>3</sub>	ethoxy	
10	CH <sub>3</sub>	CH₃	propoxy	
	CH <sub>3</sub>	СН3	1-methylethoxy	
	CH <sub>3</sub>	CH₃	n-butoxy	
	CH <sub>3</sub>	СН₃	1-methylpropoxy	
15	CH <sub>3</sub>	СН3	2-methylpropoxy	
	CH <sub>3</sub>	CH₃	1,1-dimethylethoxy	
	CH <sub>3</sub>	CH <sub>3</sub>	n-pentyloxy	
	CH <sub>3</sub>	CH <sub>3</sub>	n-hexyloxy	
20	CH <sub>3</sub>	CH <sub>3</sub>	cyclopentyl	
	CH <sub>3</sub>	CH <sub>3</sub>	cyclopentenyl	
	CH <sub>3</sub>	CH <sub>3</sub>	phenyl	

The novel active ingredients are particularly suitable for protecting various materials against degradation or destruction by bacteria or fungi or from being attacked by and covered with microorganisms. Examples of materials which can be preserved or microbicidally finished with the novel active ingredients are glues and adhesives, starch solutions, wax emulsions, clay emulsions, sizes, finishes, spinning baths, gelatine formulations, putty, joint sealants, cooling lubricants, drilling oils, fuels, plastic dispersions, emulsion paints, textiles, leather, raw hides and cosmetics. The compounds are also suitable as anti-slime agents in the paper industry, in cooling towers and in air moistening units.

The compounds I are also suitable for protecting the following 15 plant species against attack by microorganisms:

cereals (e.g., wheat, barley, rye, oats, rice, sorghum and related species); beets (e.g., sugar and fodder beets); pomes, drupes and aggregate fruit (e.g., apples, pears, plums, peaches, 20 almonds, cherries, strawberries, raspberries and blackberries); legumes (e.g., beans, lentils, peas, soybeans); oil-yielding crops (e.g., rape, mustard, poppies, olives, sunflowers, coconuts, castor-oil beans, cocoa beans, groundnuts); cucurbits (e.g., pumpkins, cucumbers, melons); fiber-yielding plants (e.g., 25 cotton, flax, hemp, jute); citrus fruit (e.g., oranges, lemons, grapefruit, tangerines); vegetables (e.g., spinach, lettuce, asparagus, cabbage varieties, carrots, onions, tomatoes, potatoes, paprika); laurel species (e.g., avocado, cinnamomum, camphor) or plants such as Indian corn, tobacco, nuts, coffee, sugar cane, 30 tea, grapes, hops, and banana and rubber trees. For the purposes of the present invention, the term "plants" is also taken to mean all types of other green growth, whether ornamentals, grassy areas, embankments, or generally low-growing cover crops.

35 For example the following microorganisms may be combatted with the novel compounds I:

Straphylococcus aureus, Escherichia coli, Klebsielle pneumoniae, Citrobacter freundii, Proteus vulgaris, Pseudomonas aeruginosa,

40 Desulfovibrio desulfuricans, Streptoverticillium rubrireticuli, Aspergillus niger, Aspergillus versicolor, Penicillium funiculosum, Penicillium expansum, Penicillium glaucum, Paecilomyces variotii, Trichoderma viride, Chaetomium globosum, Aspergillus amstelodami, Phoma pigmentovora, Phoma violacea, Aureobasidium pullulans, Saccharomyces cerevisiae, Alternaria tenuis, Stemphylium macrosporoideum, Cladosporium herbarum, Cladosporium resinae, Candida albicans, Trichophyton mentagrophytes, Geotrichum candi-

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dans, Monilia sitophila, Scenedesmus quadricauda, Chlorella vulgaris, Nostoc muscorium, Oscillatoria limosa and Anabaena constricta.

- 5 The novel substances can be converted into conventional formulations such as solutions, emulsions, suspensions, dusts, powders, pastes and granules. The application forms depend entirely on the purposes for which they are intended; they should at all events ensure a fine and uniform distribution of the active ingredient.
- 10 The formulations are produced in known manner, for example by extending the active ingredient with solvents and/or carriers, with or without the use of emulsifiers and dispersants; if water is used as solvent, it is also possible to employ other organic solvents as auxiliary solvents. Suitable auxiliaries for this
- 15 purpose are solvents such as aromatics (e.g., xylene), chlorinated aromatics (e.g., chlorobenzenes), paraffins (e.g., crude
  oil fractions), alcohols (e.g., methanol, butanol), ketones
  (e.g., cyclohexanone), amines (e.g., ethanolamine, dimethylformamide), and water; carriers such as ground natural-minerals (e.g.,
- 20 kaolins, aluminas, talc and chalk) and ground synthetic minerals (e.g., highly disperse silica and silicates); emulsifiers such as nonionic and anionic emulsifiers (e.g., polyoxyethylene fatty alcohol ethers, alkyl sulfonates and aryl sulfonates); and dispersants such as lignin-sulfite waste liquors and methylcellu-25 lose.

The fungicides generally contain from 0.1 to 95, and preferably from 0.5 to 90, wt% of active ingredient. The active ingredients are used in a purity of from 90 to 100, and preferably from 95 to 30 100, % (according to the NMR/HPLC/GC spectrum).

Usual application concentrations are - based on the weight of the material to be protected - from 0.001 to 5, and preferably from 0.01 to 2, wt% of active ingredient; when the active ingredients

35 are used for treating water, in oil production, in drilling and cutting oils, fuels, in swimming baths, cooling towers, air moistening units or in the paper industry, amounts of from 5 to 500 ppm are sufficient. Ready-to-use disinfectant solutions contain for instance from 0.5 to 10wt% of active ingredient.

40

Examples of such formulations are given below:

A solution of 90 parts by weight of compound no. 3 and 10 parts by weight of N-methyl-α-pyrrolidone, which is suitable for 45 application in the form of very fine drops.

- II. A mixture of 20 parts by weight of compound no. 4, 80 parts by weight of xylene, 10 parts by weight of the adduct of 8 to 10 moles of ethylene oxide and 1 mole of oleic acid-N-monoethanolamide, 5 parts by weight of the calcium salt of dodecylbenzenesulfonic acid, and 5 parts by weight of the adduct of 40 moles of ethylene oxide and 1 mole of castor oil. By finely dispersing the mixture in 100,000 parts by weight of water, an aqueous dispersion is obtained.
- 10 III. An aqueous dispersion of 20 parts by weight of compound no. 1, 40 parts by weight of cyclohexanone, 30 parts by weight of isobutanol, 20 parts by weight of the adduct of 40 moles of ethylene oxide and 1 mole of castor oil. A mixture of this dispersion with 100,000 parts by weight of water contains 0.02wt% of 15 the active ingredient.
- IV. An aqueous dispersion of 20 parts by weight of compound no.
  3, 25 parts by weight of cyclohexanol, 65 parts by weight of a mineral oil fraction having a boiling point between 210 and 280°C,
  20 and 10 parts by weight of the adduct of 40 moles of ethylene oxide and 1 mole of castor oil. The mixture of this dispersion with 100,000 parts by weight of water contains 0.02wt% of the active ingredient.
- 25 V. A hammer-milled mixture of 80 parts by weight of compound no. 2, 3 parts by weight of the sodium salt of disobutylnaphthalene-α-sulfonic acid, 10 parts by weight of the sodium salt of a lignin-sulfonic acid obtained from a sulfite waste liquor, and 7 parts by weight of powdered silica gel. By finely dispersing the 30 mixture in 20,000 parts by weight of water, a spray liquor containing 0.1wt% of the active ingredient is obtained.
- VI. An intimate mixture of 3 parts by weight of compound no. 1 and 97 parts by weight of particulate kaolin. The dust contains 35 3wt% of the active ingredient.
- VII. An intimate mixture of 30 parts by weight of compound no. 4, 92 parts by weight of powdered silica gel and 8 parts by weight of paraffin oil sprayed onto the surface of this silica 40 gel. This formulation of the active ingredient exhibits good adherence.
- VIII. A stable aqueous dispersion of 40 parts by weight of compound no. 2, 10 parts of the sodium salt of a phenolsulfonic 45 acid-urea-formaldehyde condensate, 2 parts of silica gel and 48 parts of water, which dispersion can be further diluted.

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IX. A stable oily dispersion of 20 parts by weight of compound no. 3, 2 parts by weight of the calcium salt of dodecylbenzene-sulfonic acid, 8 parts by weight of a fatty alcohol polyglycol ether, 2 parts by weight of the sodium salt of a phenolsulfonic 5 acid-urea-formaldehyde condensate and 68 parts by weight of a paraffinic mineral oil.

- X. A hammer-milled mixture of 10 parts by weight of compound no.

   4 parts by weight of the sodium salt of diisobutylnaphthalene-
- 10 α-sulfonic acid, 20 parts by weight of the sodium salt of a lignin-sulfonic acid obtained from a sulfite waste liquor, 38 parts by weight of silica gel, and 38 parts by weight of kaolin. By finely dispersing the mixture in 10,000 parts by weight of water, a spray liquor containing 0.1wt% of the active ingredient 15 is obtained.

Used alone, the active ingredients act as low-foaming biocides. A significant increase in the action of biocidal formulations containing these compounds is achieved if  $tri-C_6-$  to  $C_{12}-alkylmethy-$ 

20 lammonium salts, preferably in amounts of from 20 to 40wt%, based on the weight of compounds of the general formula I, are added.

The active ingredients may also be mixed with other, prior art, microbicides. In many instances, a synergistic effect is

25 achieved, i.e., the microbicidal action of the mixture is greater than the added actions of its individual components.

Prior art microbicides may be added to the novel substances in a weight ratio of from 1:100 to 100:1.

30

Examples of such active ingredients are as follows:

2-(thiocyanomethylthio)-benzothiazole

1-[2-(2,4-dichlorophenyl)-2-(2-propenyloxy)-ethyl]-1H-imidazole

35 2,4,5,6-tetrachloroisophthalodinitrile methylene bisthiocyanate

tributyltin oxide, naphthenate, benzoate, salicylate mercaptobenzothiazole

- 1,2-benzisothiazolone and its alkali metal salts
- 40 alkali metal compounds of N'-hydroxy-N-cyclohexyldiazenium oxide 2-(methoxycarbonylamino)-benzimidazole 2-methyl-3-oxo-5-chlorothiazolin-3-one trihydroxymethylnitromethane glutardialdehyde
- 45 chloroacetamide

polyhexamethylene bisguanide

5-chloro-2-methy1-4-isothiazolin-3-one + magnesium salts

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3,5-dimethyltetrahydro-1,3,5-2H-thiadiazine-2-thionehexahydrotriazine

N, N-methylolchloroacetamide

2-n-octyl-4-isothiazolin-3-one

5 oxazolidines

bisoxazolidines

2,5-dihydro-2,5-dialkoxy-2,5-dialkylfurans diethyldodecylbenzylammonium chloride

dimethyloctadecyldimethylbenzylammonium chloride

10 dimethyldidecylammonium chloride dimethyldidodecylammonium chloride trimethyltetradecylammonium chloride benzyldimethylalkyl- $(C_{12}-C_{18})$ -ammonium chloride

dichlorobenzyldimethyldodecylammonium chloride

15 cetylpyridinium chloride cetylpyridinium bromide cetyltrimethylammonium chloride laurylpyridinium chloride laurylpyridinium bisulfate

20 benzyldodecyldi(beta-oxyethyl)-ammonium chloride dodecylbenzyltrimethylammonium chloride n-alkyldimethylbenzylammonium chloride (alkyl radical: 40%  $C_{12}$ , 50%  $C_{14}$ , 10%  $C_{16}$ ) lauryldimethylethylammonium ethyl sulfate

25 n-alkyldimethyl-(1-naphthylmethyl)-ammonium chloride (alkyl radical: 98% C<sub>12</sub>, 2% C<sub>14</sub>) cetyldimethylbenzylammonium chloride lauryldimethylbenzylammonium chloride

30 Examples of further compounds which may be admixed are:

1,3-dimethylol-5,5-dimethylhydantoin dimethylolurea tetramethylolacetylenediurea

35 dimethylolglyoxalmonoureine hexamethylenetetramine glyoxal glutardialdehyde N-methylolchloroacetamide

40 1-(hydroxymethy1)-5,5-dimethylhydantoin 1,3-bis-(hydroxymethyl)-5,5-dimethylhydantoin imidazolidinylurea

1-(3-chloroally1)-3,5,7-triaza-1-azonia-adamantan chloride 1,3-bis-( $\beta$ -ethylhexyl)-5-methyl-5-amino-hexahydropyrimidine

45 1,3,5-tris-(hydroxyethyl)-1,3,5-hexahydrotriazine 1,2-dibromo-2,4-dicyanobutane 5-bromo-5-nitro-1,3-dioxane

2-bromo-2-nitropropanediol

1,1'-hexamethylene-bis-[5-(4-chlorophenyl)-biguanide]

40

4,4-diaminodiphenoxypropane

2-bromo-2-nitropropane-1,3-diol

5 sorbic acid and its salts

p-hydroxybenzoic acid and its esters and salts

zinc-2-pyridinethiol-N-oxide

2-[(hydroxylmethyl)amino]-ethanol

dithio-2,2'-bis(benzmethylamide)

10 5-chloro-2-(2,4-dichlorophenoxy)-phenol

thio-bis-(4-chlorophenol)

o-phenylphenol

chloromethyl-diiodomethylsulfone

p-chlorophenyl-3-iodopropargylformal.

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Synthesis examples

The directions given in the synthesis examples below were used, after appropriate modification of the starting materials, to obtain further compounds I. The compounds thus obtained are listed in the tables below with their physical data.

N-hydroxy-N-(2-propylphenyl)-2-chloronicotinamide

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$$H_3CH_2CH_2C$$
 $HO-N-CO$ 

30

At 0°C, 14 ml of water and 19.6 g of sodium bicarbonate are added to a solution of 15.1 g of 2-n-propylphenylhydroxylamine in 75 ml of a 2:1 mixture of ether and ligroin, and 13.6 g of 2-chloronicotinamide is then dripped in while stirring vigorously. The mix-

- 35 ture is stirred overnight at room temperature and then suction filtered. The residue is stirred for 15 minutes in 10% strength sodium bicarbonate solution, suction filtered, dissolved in ethyl acetate and dried, and the solvent is evaporated off under reduced pressure. From the crude product (14.6 g) there is iso-
- 40 lated, after recrystallization from ethanol, 12.5 g of 2-chloronicotic acid-N-hydroxy-2-n-propylanilide of m.p. 134-135°C.

Table 1

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10	Example no.	R	A	Phys. data
	1	CH (CH <sub>3</sub> ) <sub>2</sub>	2-Cl-pyridin-3-yl	107-111°C
15	2	CH <sub>2</sub> CH <sub>2</sub> CH <sub>3</sub>	2-Cl-pyridin-3-yl	134-135°C
	3	СН2СН (СН3) 2	2-Cl-pyridin-3-yl	oil
	4	phenyl	2-Cl-pyridin-3-yl	112-115°C
	5	CH <sub>2</sub> CH (CH <sub>3</sub> ) <sub>2</sub>	2-CH <sub>3</sub> , 4-CF <sub>3</sub> -thiazol-4-yl	oil
	6	phenyl	2-CH <sub>3</sub> , 4-CF <sub>3</sub> -thiazol-4-yl	173-175°C
20	7	CH <sub>2</sub> CH (CH <sub>3</sub> ) <sub>2</sub>	2,4-(CH <sub>3</sub> ) <sub>2</sub> -thiazol-4-yl	oil
	8	phenyl	$2,4-(CH_3)_2-thiazol-4-yl$	58-62°C

Examples demonstrating biological action:

Action on Botrytis cinerea

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Slices of green paprika pods were sprayed to runoff with aqueous suspensions containing (dry basis) 80% of the active ingredient and 20% of emulsifier. After the sprayed-on layer had dried, the slices were sprayed with a spore suspension [1.7·106 spores per 30 ml; 2% biomalt; water] of the fungus Botrytis cinerea and then kept for 4 days at 18°C and in high humidity.

After this period, the untreated controls exhibited 90% fungus attack, whereas the paprika slices treated with 500 ppm of compounds nos. 1 and 2 exhibited 5% attack at most.

At an application rate of 1000 ppm of compounds nos. 1 and 2 the paprika slices exhibited no attack at all, whereas the slices treated with 1000 ppm of 2-chloronicotinic acid-2-chloroanilide 40 exhibited 90% attack, just as the untreated controls.

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We claim:

1. N-Hydroxy-N-phenylcarboxamides of the formula I

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where:

15 is  $C_2-C_{12}$ -alkyl,  $C_2-C_{12}$ -alkoxy,  $C_3-C_{12}$ -alkenyl,  $C_3-C_{12}$ -alkenyloxy,  $C_3-C_6$ -alkynyl or  $C_3-C_6$ -alkynyloxy, where these groups are partially or completely halogenated;

 $C_3$ - $C_7$ -cycloalkyl,  $C_4$ - $C_7$ -cycloalkenyl,  $C_3$ - $C_7$ -cycloalkyloxy or  $C_4$ - $C_7$ -cycloalkenyloxy, where these rings may bear from one to three  $C_1$ - $C_4$ -alkyl groups;

phenyl, which may bear from one to five halogen atoms and/or from one to three of the following radicals:  $C_1-C_4-alkyl$ ,  $C_1-C_4-haloalkyl$ ,  $C_1-C_4-alkoxy$ ,  $C_1-C_4-haloalkyl$ thio;

A is a cyclic radical selected from the group of formulae Al to A7

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- 25

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$$(R^3)$$
  $R^4$   $R^6$   $R^5$   $R^6$   $R^6$ 

where:

		·

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- X is -CH<sub>2</sub>-, -S-, -SO- or -SO<sub>2</sub>-;
- Y is -O- or -S-;
- $R^1$ ,  $R^2$ ,  $R^4$ ,  $R^5$  and  $R^7$  are halogen,  $C_1-C_4$ -alkyl or  $C_1-C_4$ -haloalkyl;
- 5  $R^3$  and  $R^6$  are hydrogen, halogen or  $C_1-C_4$ -alky1;
  - n is 1 or 2, and the radicals  $R^3$  may be different when n is 2.
- N-Hydroxy-N-phenylcarboxamides of the formula I as claimed in claim 1, where R has the meanings given in claim 1 and A is a cyclic radical selected from the group having the formulae A1 to A7, where X and Y have the meanings given in claim 1 and the substituents have the following meanings:
- 15 R<sup>1</sup> is halogen, methyl or C<sub>1</sub>-haloalkyl;
  - R<sup>2</sup> is halogen or C<sub>1</sub>-haloalkyl;
  - R3 is hydrogen or methyl;
  - n is 1 or 2, and the radicals  $R^3$  may be different when n is 2;
- 20 R<sup>4</sup> is halogen or methyl;
  - R<sup>5</sup> is methyl or C<sub>1</sub>-haloalkyl;
  - R<sup>6</sup> is hydrogen, halogen or methyl;
  - R<sup>7</sup> is halogen, methyl or C<sub>1</sub>-haloalkyl.
- 25 3. N-Hydroxy-N-phenylcarboxamides of the formula I as claimed in claim 1, where R has the meanings given in claim 1 and A is cyclic radical selected from the group having the formulae A1 to A7, where X and Y have the meanings given in claim 1 and the substituents have the following meanings:

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- R1 is chloro, bromo, iodo, methyl or trifluoromethyl;
- R<sup>2</sup> is chloro or trifluoromethyl;
- R3 is hydrogen or methyl;
- n is 1 or 2, and the radicals  $R^3$  may be different when n is 2;
- R4 is chloror or methyl;
- R<sup>5</sup> is methyl, difluoromethyl or trifluoromethyl;
- R6 is hydrogen, chloro or methyl;
- R7 is chloro, methyl or trifluoromethyl.

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4. An agent for combatting injurious fungi, containing a fungicidal amount of a compound of the formula I as claimed in claim 1, 2 or 3, and inert additives.

- 5. A process for combatting injurious fungi, wherein the fungi, their habitat and/or the plants or materials to be kept free from fungi are treated with a fungicidally effective amount of a compound of the formula I as claimed in claim 1, 2 or 3.
- 6. The use of compounds I as claimed in claim 1, 2 or 3 for combatting injurious fungi.
- 7. The use of compounds I as claimed in claim 1, 2 or 3 for com-10 batting Botrytis.